

OAK RIDGE NATIONAL LABORATORY

OPERATED BY MARTIN MARIETTA ENERGY SYSTEMS, INC.

POST OFFICE BOX 2008
OAK RIDGE, TENNESSEE 37831
December 15, 1989

Mr. Richard L. Egli, Assistant Manager
Energy Research and Development
Department of Energy, Oak Ridge Operations
Post Office Box 2001
Oak Ridge, Tennessee 37831-8600

ORNL/FPO-89/121

Dear Mr. Egli:

ORNL Radioactivity Releases During 1948 and 1949

Reference: Letter from R. L. Egli to A. W. Trivelpiece, dated August 9, 1989, entitled ORNL Radioactivity Releases During 1948 and 1949

Letter from F. R. Mynatt to R. L. Egli dated, October 20, 1989, entitled ORNL Radioactivity Releases During 1948 and 1949

Enclosed is the report titled "Historic Airborne Emissions from Oak Ridge National Laboratory, 1943 to 1960" which you requested in your letter of August 9, 1989.

The original scope for the report was limited to airborne releases during 1948 and 1949. Subsequent to conversations with Wayne Hibbitts of your staff, the author expanded the scope to include airborne releases for the period of 1943 to 1960. Mr. Hibbitts believed there was a need to summarize the emissions for the entire period prior to 1960 if possible. The document referenced in your letter was not included in the review because it is still classified secret.

Sincerely,

Fred R. Mynatt

Fred R. Mynatt
Associate Director for
Chemical, Environmental, and
Health-Protection Technologies

FRM:MFT:lph

Attachment

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ChemRisk Document No. 2520

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ORNL/FPO-89/121

HISTORIC AIRBORNE EMISSIONS FROM OAK RIDGE NATIONAL LABORATORY 1943 to 1960

Oak Ridge National Laboratory
Environmental and Health Protection Division

M. F. Tardiff

December 8, 1989

This document has been approved for release
to the public by:

David R. Hamm 2/9/96
Technical Information Officer Date
ORNL Site

CAUTION

This document has not been given final patent clearance and is for internal use only. If this document is to be given public release, it must be cleared through the site Technical Information Office which will see that the proper patent and technical information reviews are completed in accordance with Energy Systems Policy.

Historic Airborne Emissions from
Oak Ridge National Laboratory
1943 to 1960

INTRODUCTION

The original mission of Oak Ridge National Laboratory (ORNL) was to construct a nuclear fission reactor for the purpose of producing sufficient amounts of plutonium to support the research and development of plutonium-uranium separation. The results of experimentation at ORNL were to be used in the design and construction of the production reactors and separations facilities at Hanford. In addition to this primary mission, ORNL was tasked with producing various radioisotopes in support of other research facilities investigating the physical, chemical, and biological properties of these materials.

The purpose of this report is to summarize the major contributors to radioactive airborne emissions during the initial operations of ORNL and during the post-war years up to 1960. The sources used for this compilation are various technical reports generated by the health physics and operations organizations during this time period. It is important to realize that the development of instrumentation and methods for the measurement and quantification of radionuclides was a new technology. For this reason, much of the earlier data consist of gross measurements. The emphasis of the documents reviewed was worker health and production efficiency. This should not be construed as a lack of concern by the people conducting the work. Similarly, little information was available on the biological effects of radiation. These early documents consistently show a concern for worker health and a willingness to modify equipment and operations as potential exposures via airborne emissions were recognized.

This report consists of four sections addressing radioactive releases to the atmosphere from the Oak Ridge Graphite Reactor (OGR), isotope separations processes, accidents, and monitoring of those releases.

AIRBORNE EMISSIONS FROM THE OGR

The OGR commenced operation on November 4, 1943. Cooling of the reactor pile was accomplished with 100,000 ft³/min of atmospheric air pulled through the core and exhausted via the 200-ft reactor stack. The neutron flux of the reactor activated stable argon in the reactor cooling air to argon-41. With the reactor operating at 3.6 MW, the discharge of argon-41 was about 470 Ci/day (1).

Production of plutonium in the reactor consisted of loading channels in the reactor core with uranium "slugs" that were enclosed in aluminum cans. Each slug weighed about 2.5 lb and had finished dimensions of 4 in. long by 1.1 in. in diameter (2). The bombardment of uranium by neutrons in the reactor core converted uranium into plutonium. The physics of this process is complex, with many isotopes being produced besides plutonium. Generally speaking, the longer the slugs resided in the core, the higher the yield of plutonium.

During the first year of operation, problems were encountered with slugs expanding and rupturing in the reactor core. Emissions associated with these events could include fission products such as radioiodines and noble gases, as well as oxides of uranium and plutonium. The composition and magnitude of the releases associated with slug ruptures is affected by many variables including: the length of time the slug was in the reactor, neutron flux at that location, temperature, and how soon the problem was detected and the slug removed. Table 1 is a listing of slug rupture events in the OGR from the start of the reactor through September 3, 1948 (3). No attempt has been made to convert these events into emission quantities.

As the Health Physics Division became aware of the presence of radioactive particulates depositing on the plant site as a consequence of slug ruptures, an aggressive campaign was initiated to determine the cause of the problem and design a solution. The concern was that, unlike gases, particulates would remain in the respiratory tract of an exposed individual, resulting in much greater health impacts. The issue of airborne particulates is first mentioned in the reviewed documents in 1948. Increased activity had been noted at the plant site associated with slug ruptures since 1943, but it wasn't until 1948 when a particulate problem was reported at Hanford that the health physicists began investigating the ORNL site for radioactive particulates. This work was initiated in May 1948 (3). By November 14, 1948, a filter house had been designed, constructed, and put in operation for the OGR stack (4). The impact of this filter house and other filter units installed on chemical processing equipment was to reduce the airborne activity by at least a factor of ten (5).

A comprehensive program to control the on-site contamination from the time previous to the filter installation was initiated as soon as the problem was identified (6).

RADIOISOTOPE SEPARATION PROCESSES

Of all the chemical processing that was conducted at ORNL, the separation of radioactive lanthanums (RALA Process) was the biggest problem with respect to gaseous and particulate emissions (7). The isotopes of interest had short half-lives. Therefore, instead of allowing short-lived fission product gases to decay away before dissolving the uranium slugs as was typical for plutonium production, the slugs were dissolved after about five days of cooling. A total of 68 RALA runs (8) were processed at ORNL before the process was discontinued. Filtration of the cell ventilation system was installed after the 28th run (5,9). Estimates of airborne particulate releases per RALA run were 6,000 mCi gamma/run prior to filtration and 3,300 mCi gamma/run after the filters were installed (10). Other production processes conducted at ORNL that had similar emission problems were Redox, iodine-131, iodine-135, and xenon-135. Apparently the length of these campaigns was short because the attention paid to them is minor when compared to the RALA process. Some information on these processes is found in references 10 and 11.

A central gas-handling system was designed to provide filtration and scrubbing for all process air streams prior to release to the atmosphere through the 250 ft stack (3039 stack). This system went online in 1950. The emissions from the OGR and the Pilot Plant (Bldg. 3019) continued to be released from their own stacks.

A study called "A Study of the Contribution of the RALA Process to Atmospheric Contamination at ORNL" (12) investigated the correlation of RALA runs to peaks of particulate activity in the facility vicinity in 1954. Figure 1 is reproduced from that report. Two main points of interest for this report are (1) there is a correlation between the RALA runs and particulate concentrations at the facility and (2) there is a significant impact upon the facility radiation signature from fallout. Ninety percent of the trappable stack effluent was iodine-131. The total trappable activity for the run analyzed averaged an activity of $3.4\text{E-}7 \text{ uCi/cm}^3$. A discrepancy between the sampler results and an ion chamber indicated that major activity components of the emissions were xenon and krypton at about $1\text{E-}3 \text{ uCi/cm}^3$. The total iodine activity released for the run was estimated to be about 30 Ci. The total emission for noble gases was estimated to be $1\text{E}5 \text{ Ci}$. No correlation was found between continuous air monitors on-site and the stack releases. It was inferred from this result that the release height of 250 ft was effective for diluting the ground level impacts of the off-gases. The major source of ground-level contamination was from the vent of a liquid-waste storage tank during jetting and sparging operations associated with the RALA run. The average activity near the waste tank vent during the RALA run was about $2\text{E-}6 \text{ uCi/cm}^3$.

Table 1. Ruptured Slug Data

Sequence Number	Row Number	Days Exposed	Date Discharged
1.	1764	84	09-27-44
2.	1264	83	10-10-44
3.	1564	26	10-31-44
4.	2165	84	12-28-44
5.	2071	169	04-08-45
6.	1770	173	04-18-45
7.	2269	200	04-23-45
8.	2373	223	04-24-45
9.	1865	238	05-04-45
10.	1563	259	05-25-45
11.	1969	258	06-20-45
12.	1772	261	07-09-45
13.	1465	160	08-06-45
14.	1865	348	09-05-45
15.	1764	30	09-10-45 =
16.	1773	91	11-13-45
17.	1867	593	12-19-45 (Donuts)
18.	2471	423	12-31-45
19.	1663	542	03-22-46
20.	1366	513	03-24-46
21.	1264	570	05-04-46
22.	1858	392	05-14-46 (T-slugs)
23.	1266	521	05-17-46
24.	2165	608	05-20-46
25.	1576	850	02-04-47
26.	1366	319	02-06-47
27.	1565	11	04-26-47
28.	2368	69	08-20-47
29.	1862	1107	10-17-47
30.	2165	141	10-31-47 (1st "W" made slug)
31.	1061	1190	11-05-47
32.	2574	1160	11-12-47
33.	2079	1160	11-30-47 (Detected-12-9-47)
34.	2074	1149	12-20-47
35.	2568	1143	12-21-47
36.	1881	1142	12-23-47
37.	2460	1204	12-26-47
38.	1871	68	01-23-48
39.	1669	72	01-27-48
40.	2165	107	03-14-48
41.	2170	110	05-02-48
42.	1159	709	05-09-48
43.	1077	1362	07-20-48

Table 1. (continued)

Sequence Number	Row Number	Days Exposed	Date Discharged
44.	961	1400	07-27-48
45.	1668	144	07-28-48
46.	1459	1420	07-29-48
47.	2874	1378	07-30-48
48.	2879	1532	07-30-48
49.	2678	1369	08-03-48
50.	1069	1404	08-31-48

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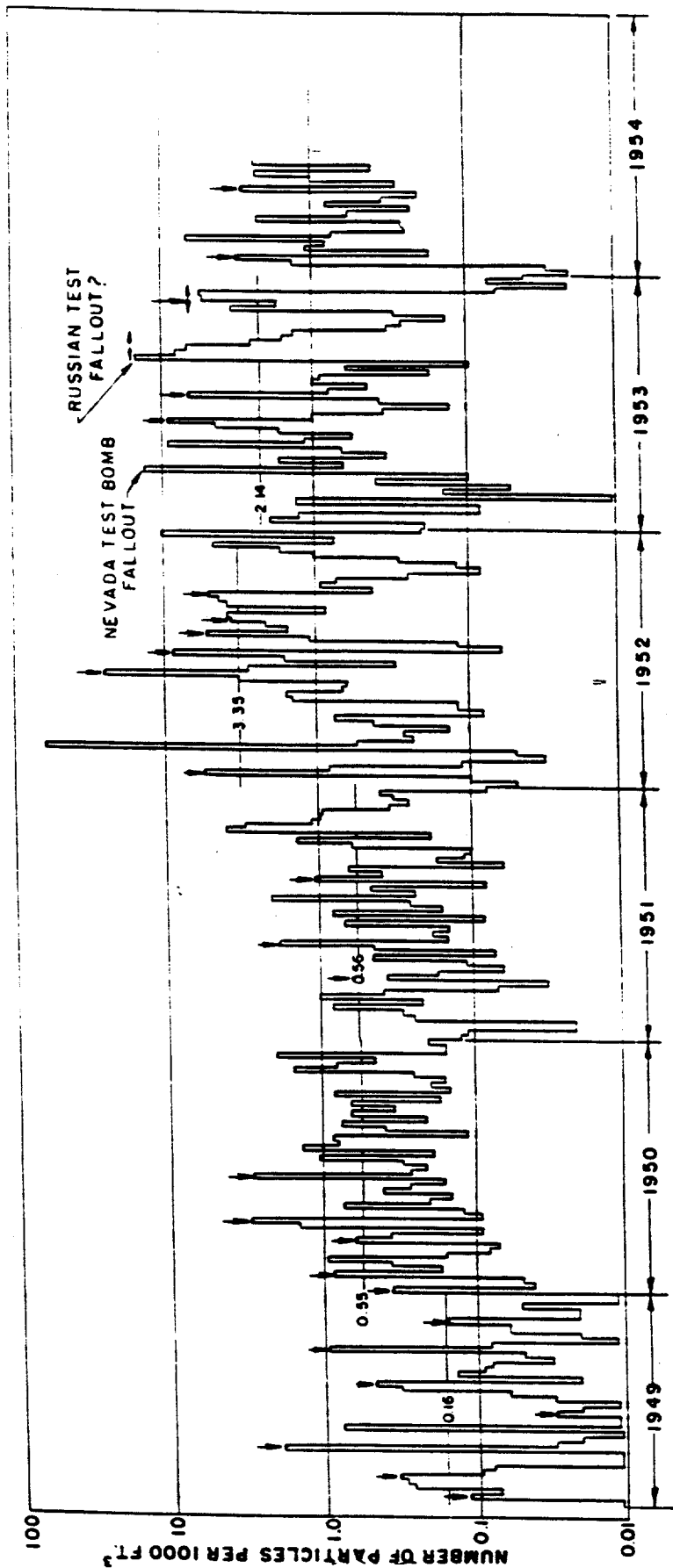


FIG. 1-AIRBORNE RADIOPARTICULATES AT ORNL
AVERAGE OF TEN MONITORS
1949-JUNE 1954 BY WEEKS

Ref. 12

ACCIDENTAL RELEASES

RALA Process

On April 29, 1954, a batch of uranium slugs was being dissolved in Building 3026-D. The batch became thermally hot between two dissolving steps, and upon the next addition of nitric acid, a violent reaction ensued. Vapors and solution were forced out of the process cell through the slug chute and solution addition lines. Air monitors in the building alarmed, and all personnel were evacuated (13). The release of activity continued for about two hours and contaminated buildings in the 3000 area (14). Exposure rates around the facilities just after the accident ranged from 5- 20 mR/h. One week later, the rates were reduced to from below background to 5 mR/h. The greatest contamination was inside the building where the release occurred. Exposure rates in this area were as high as 100 R/h (13).

Ambient air was monitored with continuous air monitors (CAMs) for submicron particles and gases which were quantified as $\mu\text{Ci}/\text{cm}^3$ of air and for larger particulates quantified as the number of particles per thousand cubic feet of air. Ambient air activity for the week including the accident averaged $2.4\text{E-}9 \mu\text{Ci}/\text{cm}^3$ for all the continuous air monitors on-site, with a maximum value of $2\text{E-}8 \mu\text{Ci}/\text{cm}^3$ at a monitor south and east of the accident area (15). A slight increase in particulate activity was also noted. The particulate activity levels associated with this event were only about 20 percent of the particulate activity found at ORNL associated with fallout from the Nevada test site and suspected Soviet weapons tests (12).

Ruthenium-106

Two short-duration releases of Ruthenium-106 occurred during the repair of central off-gas handling equipment. On November 11, 1959, a damper downstream from an exhaust fan was repaired, and a bearing was replaced on the fan motor. In the process, particles of Ruthenium-106 were knocked loose from the interior surfaces of the system. The fan was operated for about 30 minutes and then shut down at 4 p.m. In accordance with normal procedures, the fan would be put online at the beginning of the morning shift of the next day and watched during the first day of operation. When the fan was restarted for the initial 30 minutes, there was a localized release from the main stack (3039) in the direction of the east parking lot. The activity was detected by health physics personnel during routine surveys of their shoes. It was also found by an experimental stack monitor. Surveying and decontamination operations were conducted through the night. The fan was put online as scheduled at 8:15 a.m. on November 12, 1959. The shift foreman, realizing that the 3039 stack was the suspected source of contamination from the previous day, had the fan shut off at about 8:45 a.m. By 9:30 a.m., survey personnel discovered that areas that had just been cleaned were recontaminated (16). Figs. 2 and 3 show maps of the contamination resulting from these two releases. Monitoring data show both of these events to involve large diameter particulates (17). This means that the impact of the event was very localized, as shown by the isopleths in Figs. 2 and 3, and did not include a gaseous or submicron diameter component. The total Ruthenium-106 released was estimated to be from 0.3 to 15 Ci, depending upon the assumptions used in the calculations.

Plutonium-239

On November 20, 1959, a chemical explosion occurred in hot cell 6 of the Thorex Plant in Building 3019. The explosion was caused by the inadvertent mixing and heating of nitric acid with organic cleaning agents (18). The total inventory of plutonium in the cell was estimated to be 1100 g (17). Contamination was spread into the Pilot Plant, Bldg. 3019, and into the OGR. Areal contamination was localized to the area around the explosion site as shown in Fig. 4 (18).

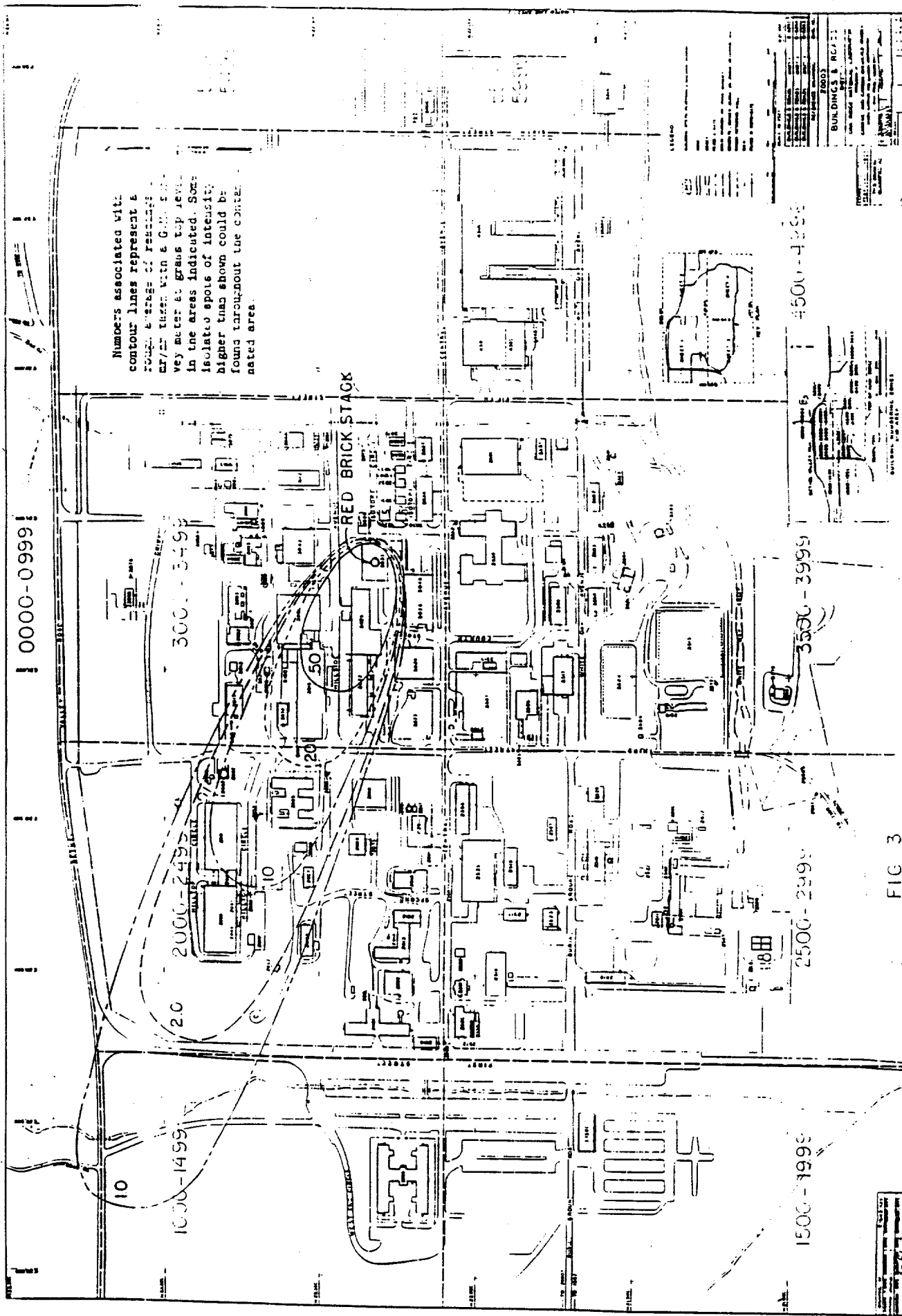
ENVIRONMENTAL MONITORING

The primary emphasis of monitoring at ORNL at the beginning of operations was personnel exposure monitoring. As noted above, there were CAMs in place by the time of the RALA accident in 1954. Information on when the CAM system was put in place and routine data reports have not been found. The average background for the area was measured in 1943 prior to operations and is reported in various sources as 0.012 mR/h (e.g. 34). Figure 5 shows the average external radiation measurements taken at the Laboratory and off-site from 1950 to 1964 (19). Figure 6 shows the average concentration of radioactive materials in air from 1956 to 1961 (20). Figure 7 shows the average fallout from 1956 to 1961 (20).

The figures show a net impact of the ORNL facility to be from 0.06 to 0.27 mR/h from 1950 to 1962. The net increase of total airborne radioactivity, as measured by CAMs, for the period of 1956 to 1960 was $25\text{E-}13$ uCi/cm³. The net impact of the site from radioactive particulates, as measured by CAMs from 1956 to 1960, was one particle per 1000 ft³ of air.

CONCLUSIONS

1. Operation of the OGR resulted in the daily production and release of 470 Ci/day of argon-41.
2. A total of 50 slug-rupture events occurred in the OGR prior to the installation of the filter house. The emission consequences are not known.
3. The production of radioactive lanthanums (RALA Process) was the major source of chronic airborne emissions from ORNL.
4. A nominal estimate of 6,000 mCi/run was made for the particulate emissions of the first 28 RALA runs. With filtration in place, the nominal release/run was reduced to 3,300 mCi of particulates.
5. A subsequent study in 1953 quantified the RALA emissions as 30 Ci of radioiodine and $1\text{E}5$ Ci of noble gases per run.
6. The primary contribution to on-site airborne activity was identified as the vent of a waste tank that received RALA liquid wastes.
7. Three accidents occurred during this time period. None of them had serious off-site consequences.
8. ORNL operations resulted in the following net impacts, as determined by routine monitoring programs:
 - External gamma at ORNL ranged from 0.06 to 0.27 mR/h above preoperational levels during 1950 - 1962.
 - Total airborne activity averaged $25\text{E-}13$ uCi/cm³ above the $10\text{E-}13$ uCi/cm³ average level found at the remote sites during 1956 - 1960.
 - Airborne particulate activity averaged 1 particle/1000 ft³ above the 1 particle/1000 ft³ average found at the remote sites during 1956 - 1960.



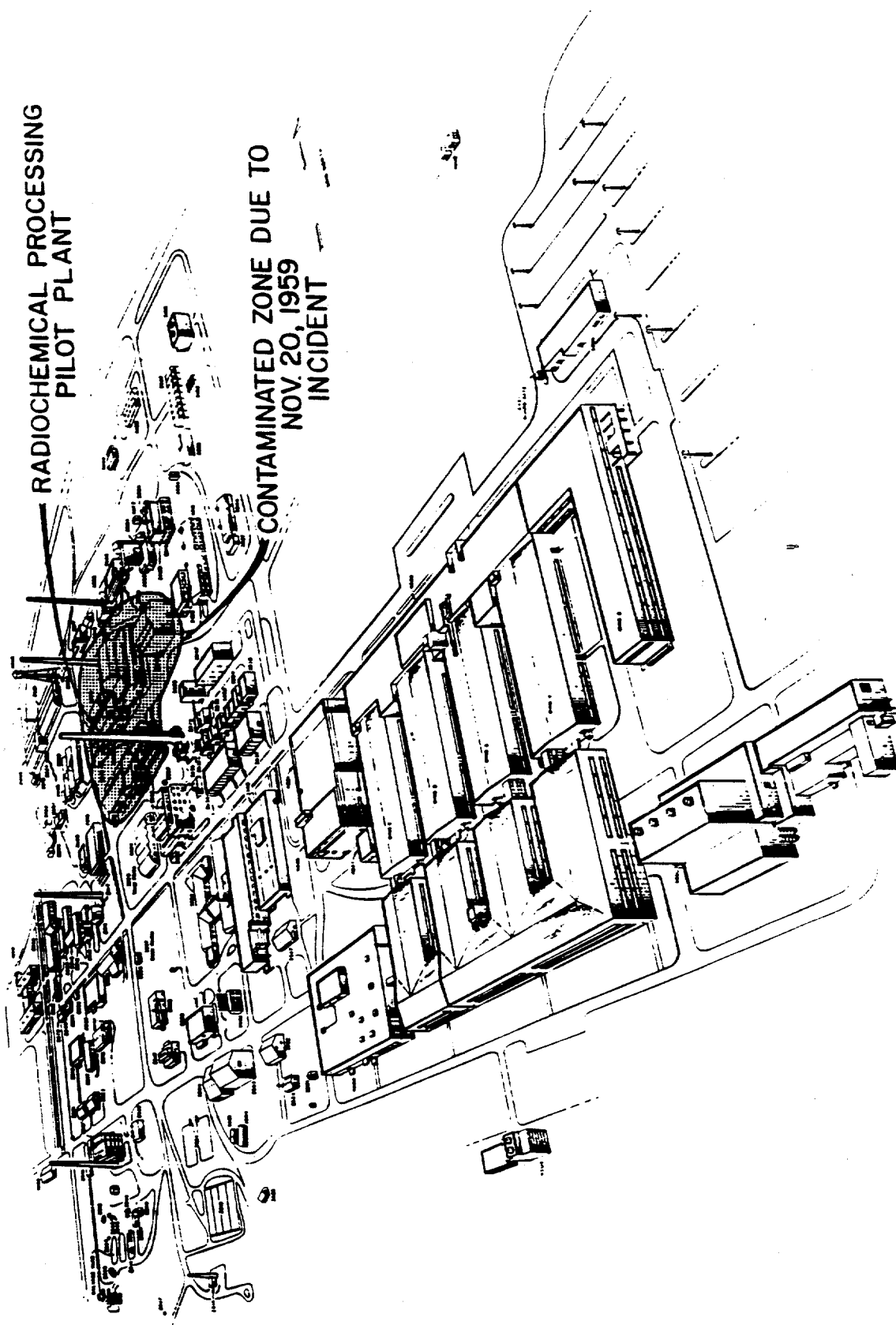


Fig. 4 Oak Ridge National Laboratory, X-10 Site.
Ref. 21

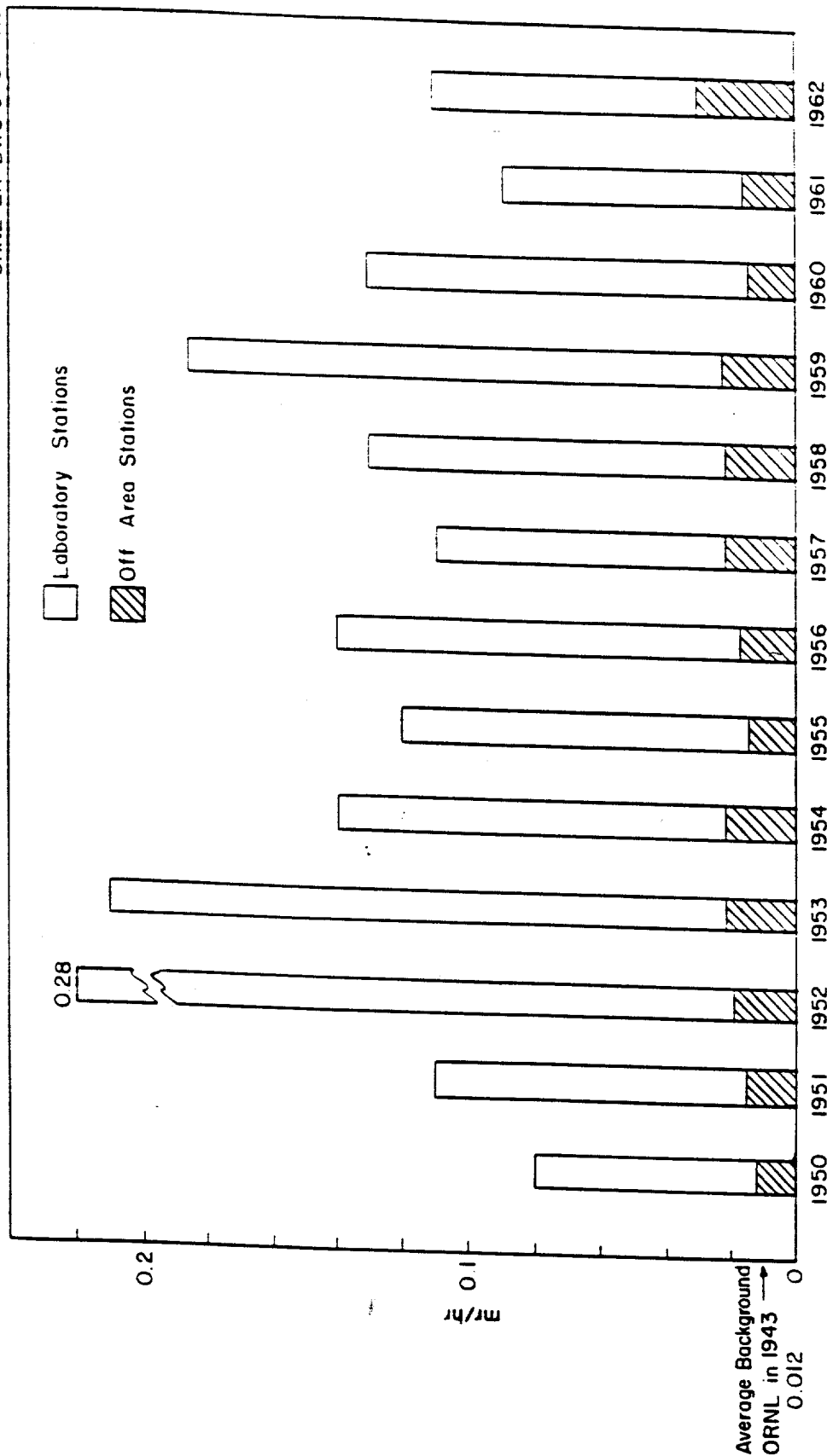


Fig. 5 Radiation Measurements Taken 3 ft Above the Ground Surface at ORNL Compared with Like Measurements Taken Elsewhere within the AEC Controlled Area for the Years 1950-1962.

Ref. 19

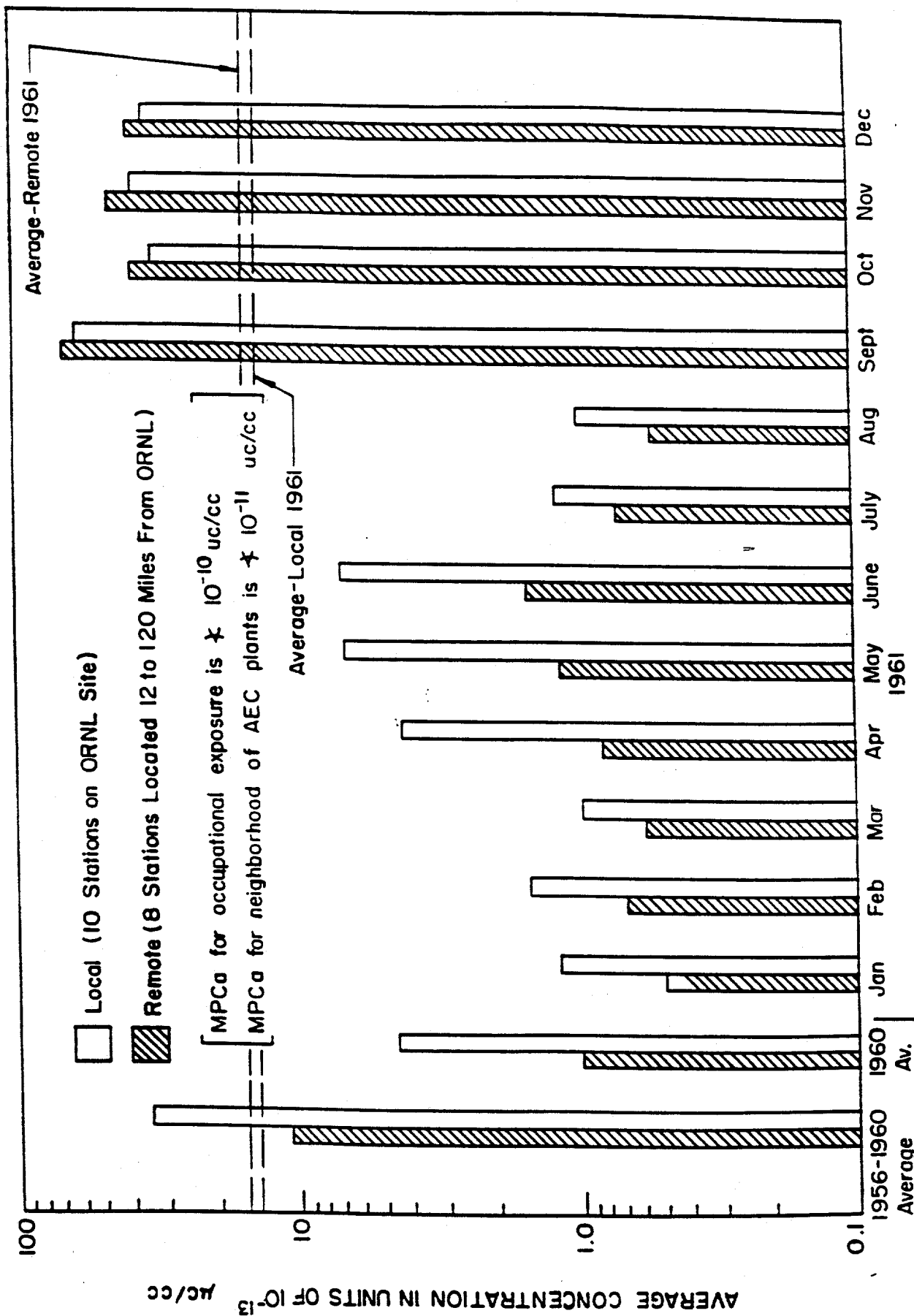


Fig. 6 Airbone Radioactivity Measurements (Filter Paper Data)

Ref. 20

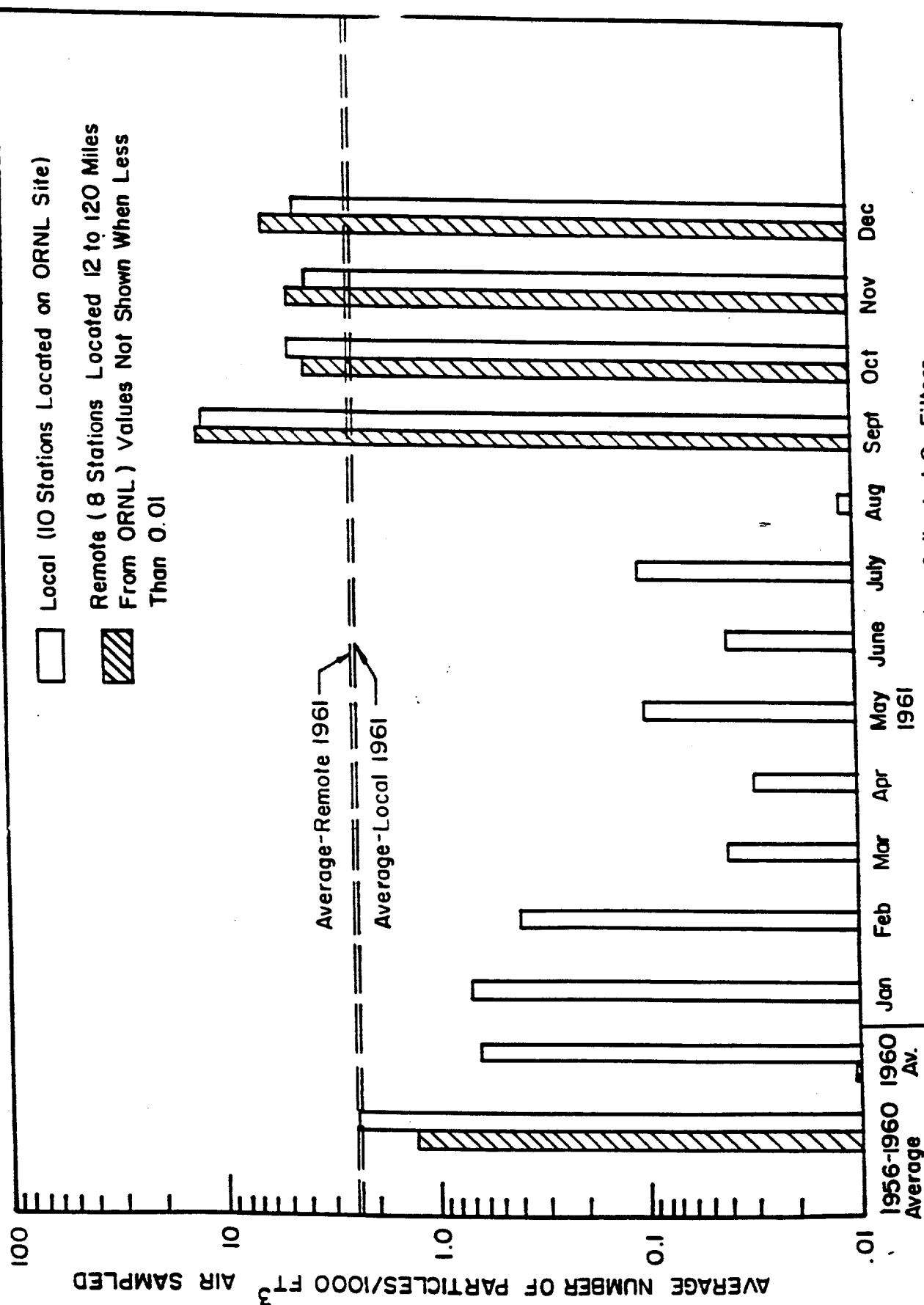


Fig. 7 Airborne Radioactive Particles Collected On Filters
(Measured by Autoradiographic Techniques)

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Internal Correspondence

MARTIN MARIETTA

MARTIN MARIETTA ENERGY SYSTEMS, INC.

Add to #2520

October 1, 1992

M. F. Tardiff

Documentation for the 1988 Letter Report

Reference: T. J. Blasing, E. Dixon, F. R. O'Donnell, W. F. Ohnesorge, and J. P. Witherspoon, "Information and Observations Regarding Early-Year Releases of Radionuclides to the Atmosphere," informal letter report, undated.

Per your verbal request, I have attempted to document the statements made in the referenced, informal letter report. This report was prepared in 1988 with a two-day deadline. Therefore, normal standards of documentation and review were not implemented.

The attached document lists the references used in the original report and, probably, a few additional ones. Copies of the cited references also are attached, unless the cited documents are classified, restricted, or contain personal information that is protected under the privacy act.

If you have any questions or require additional information, feel free to call me at your convenience.

Frank O'Donnell

F. R. O'Donnell, 4500S, MS-6102, ORNL (6-2132)

Attachment

Enclosures 33

cc: F. C. Kornegay
J. B. Murphy
File: Historical Doses

(ppp) 0252#

ATTACHMENT

Documentation prepared by F. R. O'Donnell on October 1, 1992 to substantiate findings reported in, T. J. Blasing, E. Dixon, F. R. O'Donnell, W. F. Ohnesorge, and J. P. Witherspoon, "Information and Observations Regarding Early-Year Releases of Radionuclides to the Atmosphere," informal letter report, undated.

- Paragraph 1: The historical records that show that personnel and environmental monitoring was in place at an early date include weekly area monitoring reports¹⁻⁴, area background count reports⁵⁻⁸, and weekly waste monitoring reports.⁹⁻¹²
- Paragraph 2: The maximum airborne radioactivity levels cited, 10^{-8} μCi beta/gamma and 10^{-9} μCi alpha per cubic centimeter, were taken from radiation incident reports. These records are considered confidential under the privacy act because most of them contain the names of individuals involved in and contaminated during the incidents. These reports are available for DOE inspection in Room E-255 of Building 4500S. Contacts are Elizabeth Dixon (4-7473) or Marti Devall (4-6664).
- Paragraph 3: Concentrations of ^{137}Cs in soils were taken from annual environmental monitoring reports for the years 1976-87.¹³⁻²⁴ For convenience, a recent summary of the data is included as reference 25.
- Paragraph 4: The radionuclide quantities available were estimated conservatively from references 26-29. They were obtained by using maximum reported nuclide inventories as the lower bounds and essentially doubling them to get the upper bounds).
- The original dose calculations could not be found. However, a new set of calculations is provided in reference 30.
- Paragraph 5: No documentation was found in the supporting material for this memo. The dose estimate can be supported as follows: (1) committed effective dose equivalents per curie of ^{238}Pu and ^{239}Pu released to the atmosphere during 1991 were 7.7 and 6.8 mrem/Ci, respectively; (2) 63 Ci of plutonium was recovered from slugs per year; and (3) release of all 63 Ci would result in a maximum off-site dose of 480 or 430 mrem, depending on the isotopic composition. It is doubtful that all of the processed plutonium would be released since the purpose of the project was to recover plutonium.
- Paragraph 6: The cited radionuclide release values were obtained from references 28, 29, 31, and 32. The dose estimates are reconstructed in reference 33.

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INFORMATION AND OBSERVATIONS REGARDING EARLY-YEAR RELEASES OF RADIONUCLIDES TO THE ATMOSPHERE

Contributors:

T.J. Blasing
E. Dixon
F.R. O'Donnell
W.F. Ohnesorge
J.P. Witherspoon

Based on the results of intensive, though not exhaustive, searches of archived data and reports by the contributors, it does not appear that massive releases of radionuclides to the atmosphere occurred during the early years of operations at the Oak Ridge National Laboratory. This statement is supported by historical air monitoring records, measured levels of Cs-137 in soil, and historical documents describing activities in the RaLa project, in Building 3019, and in operation of the Graphite Reactor. The records that we have examined show that personnel and environmental (air, water, laundry, etc.) monitoring were instigated very early in the history of ORNL and that the monitoring was comprehensive and well thought out.

Air monitoring records taken inside buildings following an incident indicate that, between 1948 and 1950, maximum airborne radioactivity levels were of the order of 10^{-8} microcuries beta/gamma and 10^{-9} microcuries alpha per cubic centimeter. Good records of radiation incidents exist from at least 1948. Earlier data are less formal but may be available from surveyors' logbooks. Extraction of this data will require additional time.

Measured (1976-1987) concentrations of Cs-137 in soils near and downwind of ORNL were found to be essentially the same as concentrations at other perimeter soil sampling stations and at remote sampling stations, about 1.5 picocuries per gram of soil. This finding indicates that no significantly large release of fission products from ORNL has occurred.

The RaLa project (8/45 through 10/56) was designed to recover Ba-140 and, ultimately, La-140 from fuel slugs irradiated in the Graphite Reactor and in the Hanford Reactor. Reports describing the RaLa project suggest that large quantities of radionuclides were processed. Based on data from a few documents, it appears that between 230,000 and 500,000 curies of Xe-133 and between 120,000 and 300,000 curies of I-131 were available for release to the atmosphere over the 11-year life of the project. Iodine releases undoubtedly were much less than the available amount because process scrubbers were available at an early date and because duct filters and scrubbers were installed during 1948. Release of the total available inventory of I-131 could yield an annual-average, maximum, offsite, total-body dose equivalent of less than 100 millirem. The corresponding dose equivalent for Xe-133 releases would be on the order of 1 millirem per year.

Building 3019 was used in the forties for recovery of plutonium from fuel slugs irradiated in the Graphite Reactor and in the Hanford Reactor. Fuel reprocessing technology development also was conducted in the building. As much as 1031 grams (63

curies) of Pu was recovered during a typical year. This required processing about 200 kilograms of fuel. Possible releases from this process should not give offsite dose equivalents to total body that exceed 500 millirem per year. Reprocessing development activities were of small magnitude, used fuel that was cooled for long periods, and appeared to use chemical scrubbers.

The Graphite Reactor began operation on 11/3/43. During normal conditions, about 500 curies per day of Ar-41 were released to the atmosphere. These releases should not have produced an individual offsite dose of more than 10 millirem per year. Fuel-slug failures occurred at a rate of about 13 per year (0.04% of the slugs failed). Based on the radionuclide inventory of these slugs, on the order of 550 to 1200 curies of Xe-133 and 290 to 880 curies of I-131 could have been released to the atmosphere each year. Due to the presence of filters and other devices, it is doubtful that the iodine releases reached the levels noted above. In any event, releases of the above magnitudes would not be expected to produce offsite dose equivalents approaching 500 millirem per year.

Reference 30

Dose Estimates for RaLa Releases

Over the 11-year duration of the project, we estimated that between 230,000 and 500,000 Ci of ^{133}Xe and between 120,000 and 300,000 Ci of ^{131}I were processed. Dividing these total inventories by 11 years gives annual average inventories of between 20,900 and 45,500 Ci of ^{133}Xe and between 10,900 and 27,300 Ci.

From the 1991 NESHAPS report for ORNL, we note that a release of 910 Ci of ^{133}Xe resulted in an effective dose equivalent (EDE) of 0.00012 mrem to the maximally exposed off-site individual and a release of 0.046 Ci of ^{131}I resulted in reception of 0.00030 mrem. Dividing the effective dose equivalent to the maximally exposed individual by the activity released gives the following dose factors: 0.00000013 mrem/Ci for ^{133}Xe and 0.0065 mrem/Ci for ^{131}I .

If we assume these dose factors to be applicable to RaLa releases, the maximum possible individual EDE from release of the entire annual average inventory of ^{133}Xe could be between 0.003 and 0.01 mrem/year (0.00000013 mrem/Ci x 20,900 Ci/year and 0.00000013 mrem/Ci x 45,500 Ci/year). Similarly, the maximum possible individual EDE from release of the entire annual average inventory of ^{131}I could be between 70 and 180 mrem/year (0.0065 mrem/Ci x 10,900 Ci/year and 0.0065 mrem/Ci x 27,300 Ci/year).

It is reasonable to assume release to the atmosphere of the entire ^{133}Xe inventory; but, several reports state that only a small fraction the ^{131}I inventory was released. Thus, the above range of potential doses due to releases of ^{131}I is likely a large overestimate.

Prepared: October 1, 1992
By F. R. O'Donnell, MS-6102, 4500S, ORNL (6-2132)

F.R.O.D.

Reference 33

Dose Estimates for Graphite Reactor Operational Releases

During normal operations of the Graphite Reactor, as much as 500 Ci/day of ^{41}Ar was released to the atmosphere. Such a daily release is equivalent to an annual release of 182,500 Ci. Fuel slug failures resulted in estimated releases of between 550 and 1200 Ci/year of ^{133}Xe and between 290 and 880 Ci/year of ^{131}I .

From the 1991 NESHAPS report for ORNL, we note that a release of 910 Ci of ^{133}Xe resulted in an effective dose equivalent (EDE) of 0.00012 mrem to the maximally exposed off-site individual, a release of 0.046 Ci of ^{131}I resulted in reception of 0.00030 mrem, and a release of 910 Ci of ^{41}Ar should result in a maximum EDE of 0.0043 mrem. Dividing the effective dose equivalent to the maximally exposed individual by the activity released gives the following dose factors: 0.00000013 mrem/Ci for ^{133}Xe , 0.0065 mrem/Ci for ^{131}I , and 0.0000047 mrem/Ci for ^{41}Ar .

If we assume these dose factors to be applicable to Graphite Reactor releases, the maximum possible individual EDE from the annual release of ^{133}Xe could be between 0.000073 and 0.0001 mrem/year (0.00000013 mrem/Ci x 550 Ci/year and 0.00000013 mrem/Ci x 1200 Ci/year). Similarly, the maximum possible individual EDE from the annual release of ^{131}I could be between 2 and 6 mrem/year (0.0065 mrem/Ci x 550 Ci/year and 0.0065 mrem/Ci x 880 Ci/year). The maximum possible individual EDE from an annual release of 182,500 Ci of ^{41}Ar could be about 0.9 mrem/year (0.0000047 mrem/Ci x 182,500 Ci/year).

Prepared: October 1, 1992

By F. R. O'Donnell, MS-6102, 4500S, ORNL (6-2132)

F.R.O.D.

290 = 22 Ci I-131 per slug
880 = 68 Ci I-131 per slug
550 = 42 Ci Xe-133 per slug
1200 = 92 Ci Xe-133 per slug



STATE OF TENNESSEE
CORDELL HULL BUILDING
DEPARTMENT OF HEALTH
NASHVILLE, TENNESSEE 37247

MEMORANDUM

Date: October 27, 1992

To: Steve Ripple

From: Mary Yarbrough *MY*

Re: Information related to I-131 and Plutonium releases

In follow-up to our phone conversation on October 14, 1992, I wanted to pass on to you that Wayne Hibbitts had mentioned that DOE was in the process of documenting the dose estimates made as a result of I-131 and Plutonium releases in the early years of the Oak Ridge facilities operations. He has references for both of these estimates and has said he will pass this on to my division. However, I think it would be wise for Tom Widner to discuss this with Mr. Hibbitts personally. While I am aware of the I-131 dose estimates made by DOE in 1988, I first heard of the estimates for Plutonium in this conversation with him.

Also, if I understood you correctly, Mr. Hibbitts has also told ChemRisk that he is aware of information regarding non-radioactive dose estimates that DOE estimated in recent years. I would appreciate knowing more about that when you have had a chance to review the information.

Thanks



Department of Energy

Oak Ridge Field Office

P.O. Box 2001

Oak Ridge, Tennessee 37831—

October 15, 1992



Dr. Mary I. Yarbrough, Director
Division of Environmental Epidemiology
Tennessee Department of Health
Cordell Hull Building
Nashville, Tennessee 37247-4912

Dear Dr. Yarbrough:

HEALTH ASSESSMENT STEERING PANEL REQUEST

At the October 5 meeting of the Panel you asked that I provide you a copy of the 1988 informal paper that Ralph Hutcheson asked me about during the meeting. The paper was prepared on a "crash" basis for use during a news conference in 1988 covering issuance of the historical rad release report and our annual site environmental report. The authors did not document their calculations, or their references. As a result of a verbal request by Steven Smith of the Oak Ridge Environmental Peace Alliance (OREPA), we asked ORNL to review the paper and provide backup information for OREPA's use and for placement in our public Reading Room.

The informal paper and supporting references, which include calculations that were recently recreated by Frank O'Donnell, are enclosed. I am now working with the DOE-ORNL Site Office and ORNL staff in an effort to assure that the backup information adequately supports the informal paper. I have asked that the following additional information be provided.

- (1) More detail on how the assumed source terms were arrived at for RaLa and the Graphite Reactor.
- (2) The basis for the statement in the report that "process scrubbers were available at an early date...".
- (3) O'Donnell indicated that one document he came across is still classified (ORNL-170). In order to make it available, the ORNL Site Office has initiated declassification so it can be placed in the Reading Room.
- (4) O'Donnell also reviewed a file of incident reports that contain Privacy Act protected information. I have asked that all documents in the file be redacted so they can also be placed in the Reading Room. The site office is determining the feasibility of doing so.
- (5) The doses generated are committed effective dose equivalents (EDE). O'Donnell agreed to review the computer program outputs and, if available, provide thyroid doses for the iodine releases. He does not think infant thyroid dose data is included, but that adult doses may be. He stated that the dose calculational program is the one

Dr. Mary I. Yarbrough

-2-

October 15, 1992

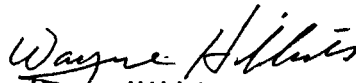
prescribed by the USEPA for Rad NESHAP compliance. EPA's standards are now for EDEs; thus, he thinks their computer program may be limited to EDE rather than being organ specific.

At the Nashville meeting I indicated that I was attempting to obtain a peer review of this informal paper. ORNL staff have since convinced me that the nature of the paper does not warrant a peer review. The paper does not represent a dose reconstruction, since the authors did not believe that sufficient information was available to provide realistic estimates of releases and thus produce realistic public radiation dose estimates. (I make this statement in reference to the RaLa and Pu recovery source terms. The Graphite Reactor source term range may be realistic; I do not yet understand the basis for the range.) The authors of the paper used readily available information and assumed very large releases of iodine and plutonium in an effort to make it very unlikely that they would underestimate public doses. They then ratioed the assumed source terms with 1987 annual source terms/dose calculations. There was neither sufficient time nor information available to make calculations for short term releases. The lack of time also resulted in the need to use current release points, rather than attempting to tailor the calculation for other points.

I should also point out that this paper focused on what the authors believed to be the most significant sources of airborne releases. Further study of the history of ORNL operations may discover additional releases of interest.

I have taken the liberty to enclose an additional document. It is a December 8, 1989, report entitled "Historical Airborne Emissions From Oak Ridge National Laboratory 1943 to 1960." It was placed in our Reading Room in early September of this year, along with its references, in response to the same verbal request from Steven Smith. A summary of this 1989 report was included in our annual site environmental report covering the year 1989 (the annual report was issued in 1990).

Sincerely,



H. Wayne Hibbitts
Deputy Assistant Manager
for Environment, Safety, and Quality

Enclosures

Wayne - your copy.

lcy-Dave Howard, SE-33
Margaret Wilson, SE-31
Linda McLaren, RLWMD

OAK RIDGE NATIONAL LABORATORY

OPERATED BY MARTIN MARIETTA ENERGY SYSTEMS, INC.

POST OFFICE BOX 2008
OAK RIDGE, TENNESSEE 37831
December 15, 1989

Mr. Richard L. Egli, Assistant Manager
Energy Research and Development
Department of Energy, Oak Ridge Operations
Post Office Box 2001
Oak Ridge, Tennessee 37831-8600

Dear Mr. Egli:

ORNL Radioactivity Releases During 1948 and 1949

Reference: Letter from R. L. Egli to A. W. Trivelpiece, dated August 9, 1989, entitled ORNL Radioactivity Releases During 1948 and 1949

Letter from F. R. Mynatt to R. L. Egli dated, October 20, 1989, entitled ORNL Radioactivity Releases During 1948 and 1949

Enclosed is the report titled "Historic Airborne Emissions from Oak Ridge National Laboratory, 1943 to 1960" which you requested in your letter of August 9, 1989.

The original scope for the report was limited to airborne releases during 1948 and 1949. Subsequent to conversations with Wayne Hibbitts of your staff, the author expanded the scope to include airborne releases for the period of 1943 to 1960. Mr. Hibbitts believed there was a need to summarize the emissions for the entire period prior to 1960 if possible. The document referenced in your letter was not included in the review because it is still classified secret.

Sincerely,

Fred R. Mynatt

Fred R. Mynatt
Associate Director for
Chemical, Environmental, and
Health-Protection Technologies

FRM:MFT:lph

Attachment

cc w/att: R. N. Collier, DOE/ORO
H. W. Hibbitts, SE-30, ORO
L. T. Radcliffe, ER-12, ORO
P. S. Rohwer
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T. H. Row

J. H. Swanks
M. F. Tardiff
A. W. Trivelpiece
D. Underwood, ER-121, ORO
File - RC

HISTORIC AIRBORNE EMISSIONS FROM
OAK RIDGE NATIONAL LABORATORY
1943 to 1960

Oak Ridge National Laboratory
Environmental and Health Protection Division

M. F. Tardiff

December 8, 1989

CAUTION

This document has not been given final patent clearance and is for internal use only. If this document is to be given public release, it must be cleared through the site Technical Information Office which will see that the proper patent and technical information reviews are completed in accordance with Energy Systems Policy.

Historic Airborne Emissions from
Oak Ridge National Laboratory
1943 to 1960

INTRODUCTION

The original mission of Oak Ridge National Laboratory (ORNL) was to construct a nuclear fission reactor for the purpose of producing sufficient amounts of plutonium to support the research and development of plutonium-uranium separation. The results of experimentation at ORNL were to be used in the design and construction of the production reactors and separations facilities at Hanford. In addition to this primary mission, ORNL was tasked with producing various radioisotopes in support of other research facilities investigating the physical, chemical, and biological properties of these materials.

The purpose of this report is to summarize the major contributors to radioactive airborne emissions during the initial operations of ORNL and during the post-war years up to 1960. The sources used for this compilation are various technical reports generated by the health physics and operations organizations during this time period. It is important to realize that the development of instrumentation and methods for the measurement and quantification of radionuclides was a new technology. For this reason, much of the earlier data consist of gross measurements. The emphasis of the documents reviewed was worker health and production efficiency. This should not be construed as a lack of concern by the people conducting the work. Similarly, little information was available on the biological effects of radiation. These early documents consistently show a concern for worker health and a willingness to modify equipment and operations as potential exposures via airborne emissions were recognized.

This report consists of four sections addressing radioactive releases to the atmosphere from the Oak Ridge Graphite Reactor (OGR), isotope separations processes, accidents, and monitoring of those releases.

AIRBORNE EMISSIONS FROM THE OGR

The OGR commenced operation on November 4, 1943. Cooling of the reactor pile was accomplished with 100,000 ft³/min of atmospheric air pulled through the core and exhausted via the 200-ft reactor stack. The neutron flux of the reactor activated stable argon in the reactor cooling air to argon-41. With the reactor operating at 3.6 MW, the discharge of argon-41 was about 470 Ci/day (1).

Production of plutonium in the reactor consisted of loading channels in the reactor core with uranium "slugs" that were enclosed in aluminum cans. Each slug weighed about 2.5 lb and had finished dimensions of 4 in. long by 1.1 in. in diameter (2). The bombardment of uranium by neutrons in the reactor core converted uranium into plutonium. The physics of this process is complex, with many isotopes being produced besides plutonium. Generally speaking, the longer the slugs resided in the core, the higher the yield of plutonium.

During the first year of operation, problems were encountered with slugs expanding and rupturing in the reactor core. Emissions associated with these events could include fission products such as radioiodines and noble gases, as well as oxides of uranium and plutonium. The composition and magnitude of the releases associated with slug ruptures is affected by many variables including: the length of time the slug was in the reactor, neutron flux at that location, temperature, and how soon the problem was detected and the slug removed. Table 1 is a listing of slug rupture events in the OGR from the start of the reactor through September 3, 1948 (3). No attempt has been made to convert these events into emission quantities.

As the Health Physics Division became aware of the presence of radioactive particulates depositing on the plant site as a consequence of slug ruptures, an aggressive campaign was initiated to determine the cause of the problem and design a solution. The concern was that, unlike gases, particulates would remain in the respiratory tract of an exposed individual, resulting in much greater health impacts. The issue of airborne particulates is first mentioned in the reviewed documents in 1948. Increased activity had been noted at the plant site associated with slug ruptures since 1943, but it wasn't until 1948 when a particulate problem was reported at Hanford that the health physicists began investigating the ORNL site for radioactive particulates. This work was initiated in May 1948 (3). By November 14, 1948, a filter house had been designed, constructed, and put in operation for the OGR stack (4). The impact of this filter house and other filter units installed on chemical processing equipment was to reduce the airborne activity by at least a factor of ten (5).

A comprehensive program to control the on-site contamination from the time previous to the filter installation was initiated as soon as the problem was identified (6).

RADIOISOTOPE SEPARATION PROCESSES

Of all the chemical processing that was conducted at ORNL, the separation of radioactive lanthanums (RALA Process) was the biggest problem with respect to gaseous and particulate emissions (7). The isotopes of interest had short half-lives. Therefore, instead of allowing short-lived fission product gases to decay away before dissolving the uranium slugs as was typical for plutonium production, the slugs were dissolved after about five days of cooling. A total of 68 RALA runs (8) were processed at ORNL before the process was discontinued. Filtration of the cell ventilation system was installed after the 28th run (5,9). Estimates of airborne particulate releases per RALA run were 6,000 mCi gamma/run prior to filtration and 3,300 mCi gamma/run after the filters were installed (10). Other production processes conducted at ORNL that had similar emission problems were Redox, iodine-131, iodine-135, and xenon-135. Apparently the length of these campaigns was short because the attention paid to them is minor when compared to the RALA process. Some information on these processes is found in references 10 and 11.

A central gas-handling system was designed to provide filtration and scrubbing for all process air streams prior to release to the atmosphere through the 250 ft stack (3039 stack). This system went online in 1950. The emissions from the OGR and the Pilot Plant (Bldg. 3019) continued to be released from their own stacks.

A study called "A Study of the Contribution of the RALA Process to Atmospheric Contamination at ORNL" (12) investigated the correlation of RALA runs to peaks of particulate activity in the facility vicinity in 1954. Figure 1 is reproduced from that report. Two main points of interest for this report are (1) there is a correlation between the RALA runs and particulate concentrations at the facility and (2) there is a significant impact upon the facility radiation signature from fallout. Ninety percent of the trappable stack effluent was iodine-131. The total trappable activity for the run analyzed averaged an activity of $3.4\text{E-}7 \text{ uCi/cm}^3$. A discrepancy between the sampler results and an ion chamber indicated that major activity components of the emissions were xenon and krypton at about $1\text{E-}3 \text{ uCi/cm}^3$. The total iodine activity released for the run was estimated to be about 30 Ci. The total emission for noble gases was estimated to be $1\text{E}5 \text{ Ci}$. No correlation was found between continuous air monitors on-site and the stack releases. It was inferred from this result that the release height of 250 ft was effective for diluting the ground level impacts of the off-gases. The major source of ground-level contamination was from the vent of a liquid-waste storage tank during jetting and sparging operations associated with the RALA run. The average activity near the waste tank vent during the RALA run was about $2\text{E-}6 \text{ uCi/cm}^3$.

Table 1. Ruptured Slug Data

Sequence Number	Row Number	Days Exposed	Date Discharged
1.	1764	84	09-27-44
2.	1264	83	10-10-44
3.	1564	26	10-31-44
4.	2165	84	12-28-44
5.	2071	169	04-08-45
6.	1770	173	04-18-45
7.	2269	200	04-23-45
8.	2373	223	04-24-45
9.	1865	238	05-04-45
10.	1563	259	05-25-45
11.	1969	258	06-20-45
12.	1772	261	07-09-45
13.	1465	160	08-06-45
14.	1865	348	09-05-45
15.	1764	30	09-10-45
16.	1773	91	11-13-45
17.	1867	593	12-19-45 (Donuts)
18.	2471	423	12-31-45
19.	1663	542	03-22-46
20.	1366	513	03-24-46
21.	1264	570	05-04-46
22.	1858	392	05-14-46 (T-slugs)
23.	1266	521	05-17-46
24.	2165	608	05-20-46
25.	1576	850	02-04-47
26.	1366	319	02-06-47
27.	1565	11	04-26-47
28.	2368	69	08-20-47
29.	1862	1107	10-17-47
30.	2165	141	10-31-47 (1st "W" made slug)
31.	1061	1190	11-05-47
32.	2574	1160	11-12-47
33.	2079	1160	11-30-47 (Detected-12-9-47)
34.	2074	1149	12-20-47
35.	2568	1143	12-21-47
36.	1881	1142	12-23-47
37.	2460	1204	12-26-47
38.	1871	68	01-23-48
39.	1669	72	01-27-48
40.	2165	107	03-14-48
41.	2170	110	05-02-48
42.	1159	709	05-09-48
43.	1077	1362	07-20-48

Table 1. (continued)

Sequence Number	Row Number	Days Exposed	Date Discharged
44.	961	1400	07-27-48
45.	1668	144	07-28-48
46.	1459	1420	07-29-48
47.	2874	1378	07-30-48
48.	2879	1532	07-30-48
49.	2678	1369	08-03-48
50.	1069	1404	08-31-48

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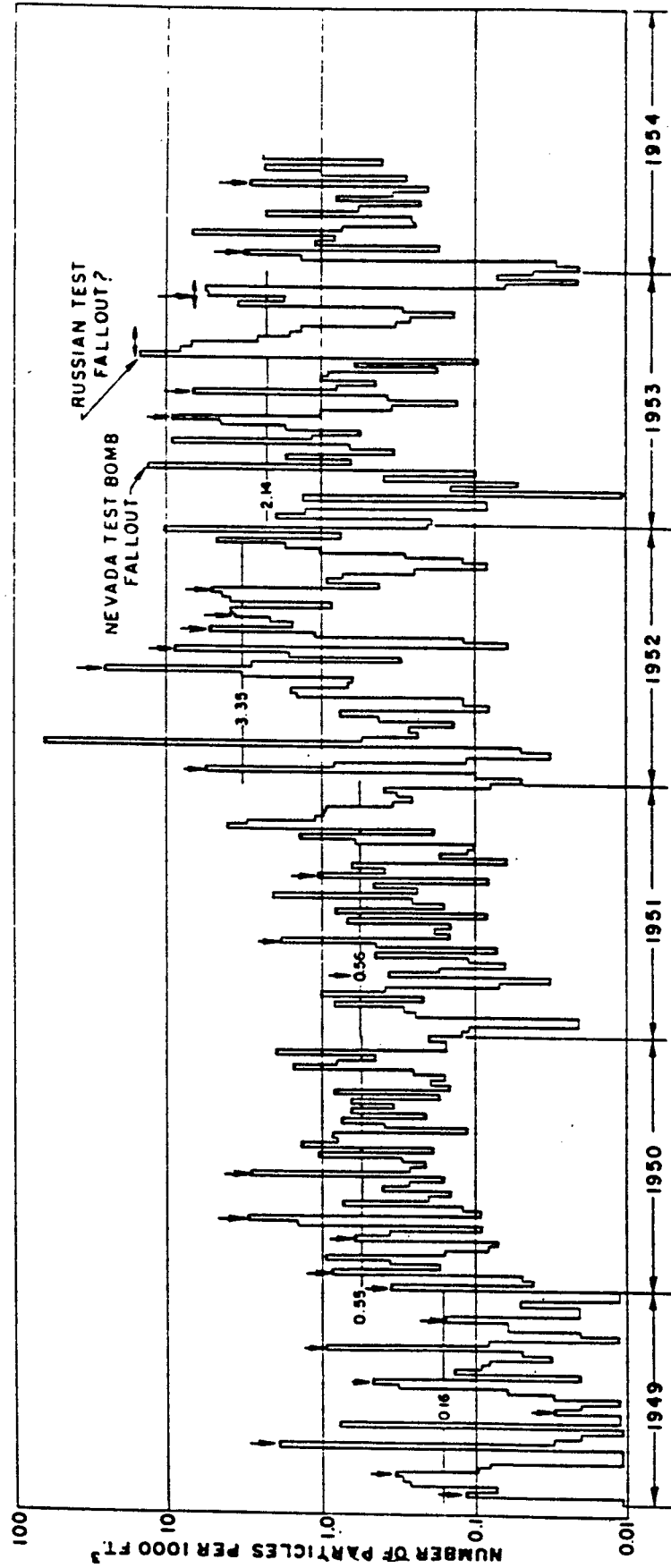


FIG.1-AIRBORNE RADIOPARTICULATES AT ORNL
AVERAGE OF TEN MONITORS
1949-JUNE 1954 BY WEEKS
Ref. 12

ACCIDENTAL RELEASES

RALA Process

On April 29, 1954, a batch of uranium slugs was being dissolved in Building 3026-D. The batch became thermally hot between two dissolving steps, and upon the next addition of nitric acid, a violent reaction ensued. Vapors and solution were forced out of the process cell through the slug chute and solution addition lines. Air monitors in the building alarmed, and all personnel were evacuated (13). The release of activity continued for about two hours and contaminated buildings in the 3000 area (14). Exposure rates around the facilities just after the accident ranged from 5- 20 mR/h. One week later, the rates were reduced to from below background to 5 mR/h. The greatest contamination was inside the building where the release occurred. Exposure rates in this area were as high as 100 R/h (13).

Ambient air was monitored with continuous air monitors (CAMs) for submicron particles and gases which were quantified as $\mu\text{Ci}/\text{cm}^3$ of air and for larger particulates quantified as the number of particles per thousand cubic feet of air. Ambient air activity for the week including the accident averaged $2.4\text{E}-9 \mu\text{Ci}/\text{cm}^3$ for all the continuous air monitors on-site, with a maximum value of $2\text{E}-8 \mu\text{Ci}/\text{cm}^3$ at a monitor south and east of the accident area (15). A slight increase in particulate activity was also noted. The particulate activity levels associated with this event were only about 20 percent of the particulate activity found at ORNL associated with fallout from the Nevada test site and suspected Soviet weapons tests (12).

Ruthenium-106

Two short-duration releases of Ruthenium-106 occurred during the repair of central off-gas handling equipment. On November 11, 1959, a damper downstream from an exhaust fan was repaired, and a bearing was replaced on the fan motor. In the process, particles of Ruthenium-106 were knocked loose from the interior surfaces of the system. The fan was operated for about 30 minutes and then shut down at 4 p.m. In accordance with normal procedures, the fan would be put online at the beginning of the morning shift of the next day and watched during the first day of operation. When the fan was restarted for the initial 30 minutes, there was a localized release from the main stack (3039) in the direction of the east parking lot. The activity was detected by health physics personnel during routine surveys of their shoes. It was also found by an experimental stack monitor. Surveying and decontamination operations were conducted through the night. The fan was put online as scheduled at 8:15 a.m. on November 12, 1959. The shift foreman, realizing that the 3039 stack was the suspected source of contamination from the previous day, had the fan shut off at about 8:45 a.m. By 9:30 a.m., survey personnel discovered that areas that had just been cleaned were recontaminated (16). Figs. 2 and 3 show maps of the contamination resulting from these two releases. Monitoring data show both of these events to involve large diameter particulates (17). This means that the impact of the event was very localized, as shown by the isopleths in Figs. 2 and 3, and did not include a gaseous or submicron diameter component. The total Ruthenium-106 released was estimated to be from 0.3 to 15 Ci, depending upon the assumptions used in the calculations.

Plutonium-239

On November 20, 1959, a chemical explosion occurred in hot cell 6 of the Thorex Plant in Building 3019. The explosion was caused by the inadvertent mixing and heating of nitric acid with organic cleaning agents (18). The total inventory of plutonium in the cell was estimated to be 1100 g (17). Contamination was spread into the Pilot Plant, Bldg. 3019, and into the OGR. Areal contamination was localized to the area around the explosion site as shown in Fig. 4 (18).

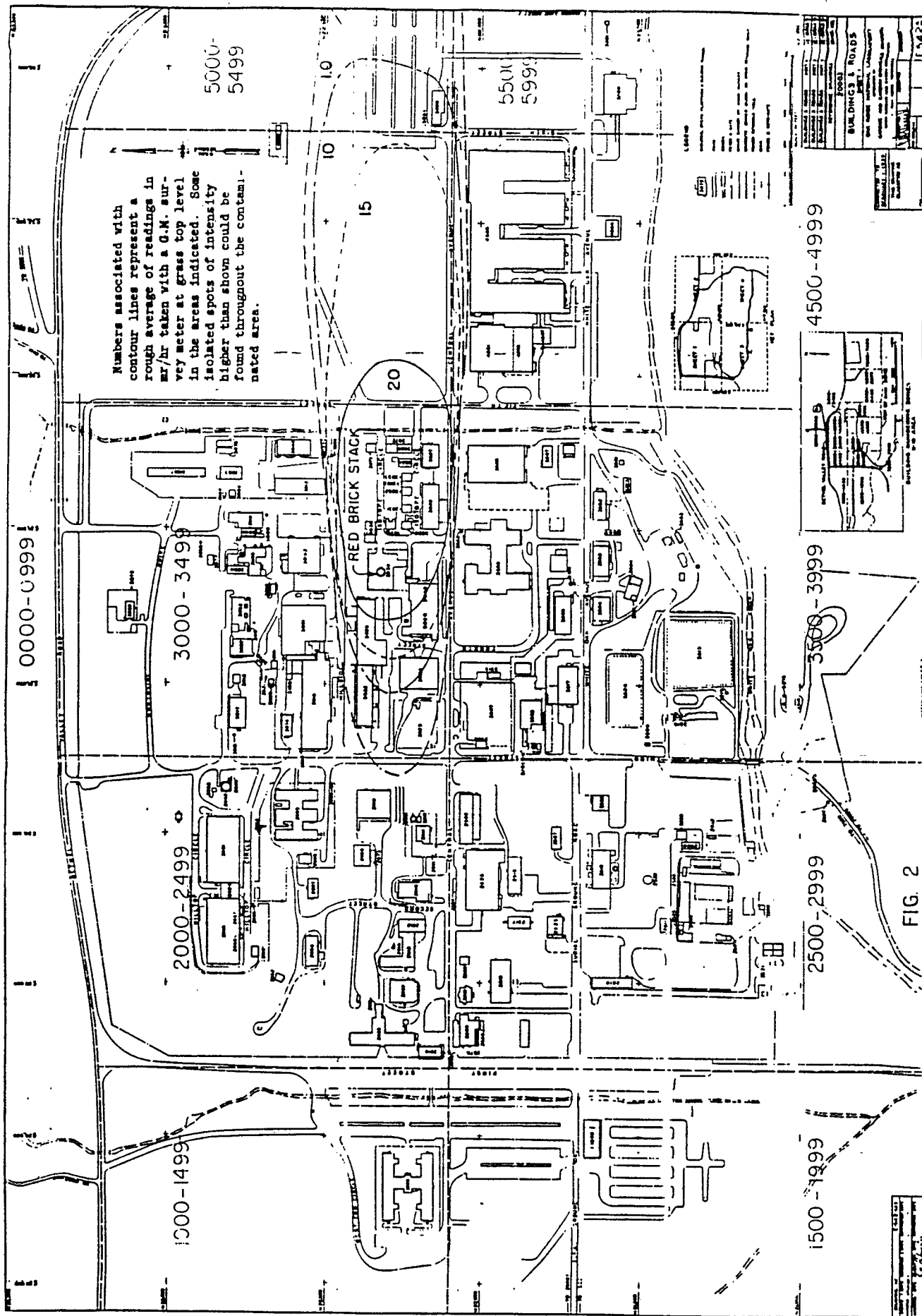
ENVIRONMENTAL MONITORING

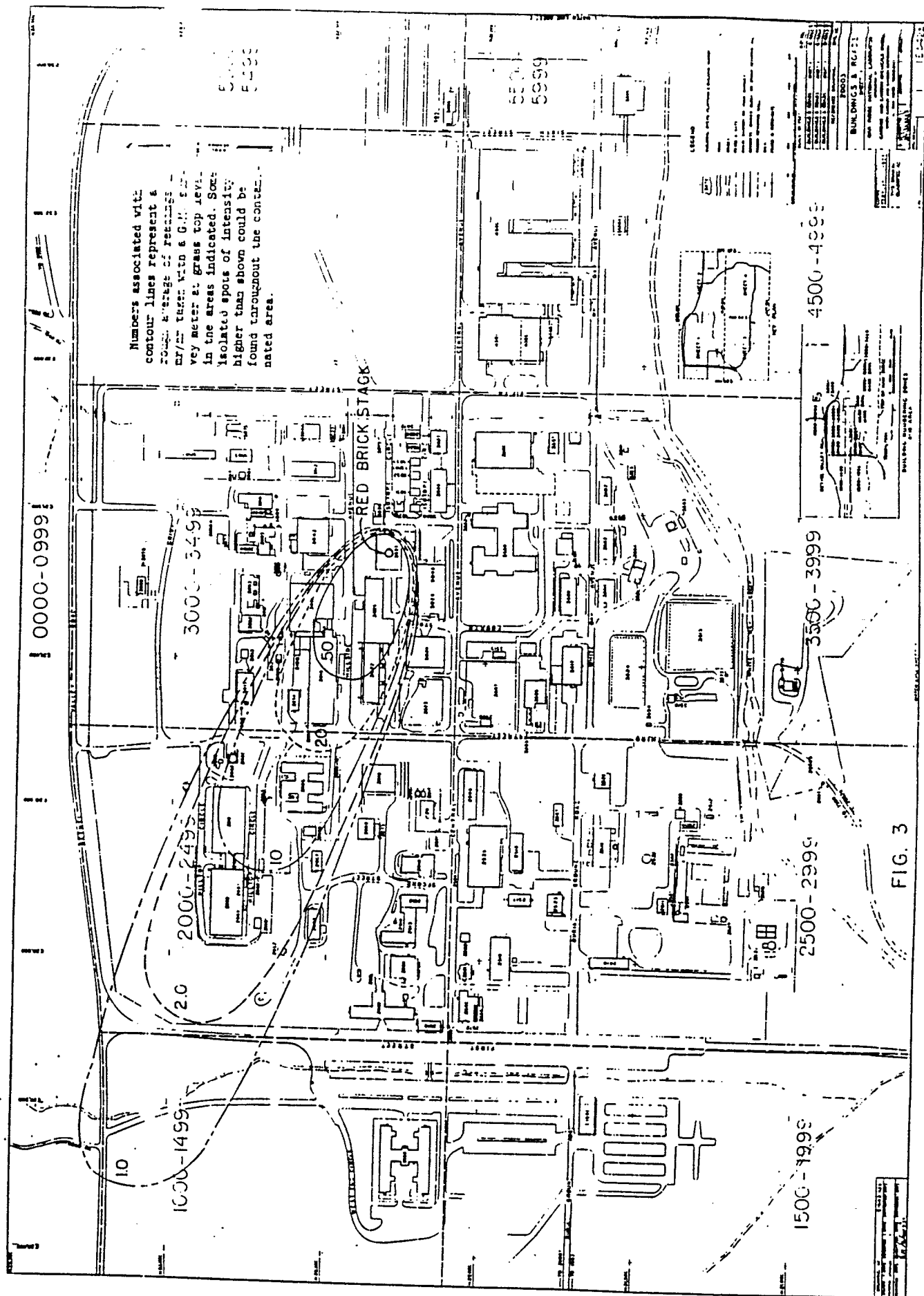
The primary emphasis of monitoring at ORNL at the beginning of operations was personnel exposure monitoring. As noted above, there were CAMs in place by the time of the RALA accident in 1954. Information on when the CAM system was put in place and routine data reports have not been found. The average background for the area was measured in 1943 prior to operations and is reported in various sources as 0.012 mR/h (e.g. 34). Figure 5 shows the average external radiation measurements taken at the Laboratory and off-site from 1950 to 1964 (19). Figure 6 shows the average concentration of radioactive materials in air from 1956 to 1961 (20). Figure 7 shows the average fallout from 1956 to 1961 (20).

The figures show a net impact of the ORNL facility to be from 0.06 to 0.27 mR/h from 1950 to 1962. The net increase of total airborne radioactivity, as measured by CAMs, for the period of 1956 to 1960 was $25\text{E-}13$ uCi/cm³. The net impact of the site from radioactive particulates, as measured by CAMs from 1956 to 1960, was one particle per 1000 ft³ of air.

CONCLUSIONS

1. Operation of the OGR resulted in the daily production and release of 470 Ci/day of argon-41.
2. A total of 50 slug-rupture events occurred in the OGR prior to the installation of the filter house. The emission consequences are not known.
3. The production of radioactive lanthanums (RALA Process) was the major source of chronic airborne emissions from ORNL.
4. A nominal estimate of 6,000 mCi/run was made for the particulate emissions of the first 28 RALA runs. With filtration in place, the nominal release/run was reduced to 3,300 mCi of particulates.
5. A subsequent study in 1953 quantified the RALA emissions as 30 Ci of radioiodine and $1\text{E}5$ Ci of noble gases per run.
6. The primary contribution to on-site airborne activity was identified as the vent of a waste tank that received RALA liquid wastes.
7. Three accidents occurred during this time period. None of them had serious off-site consequences.
8. ORNL operations resulted in the following net impacts, as determined by routine monitoring programs:
 - External gamma at ORNL ranged from 0.06 to 0.27 mR/h above preoperational levels during 1950 - 1962.
 - Total airborne activity averaged $25\text{E-}13$ uCi/cm³ above the $10\text{E-}13$ uCi/cm³ average level found at the remote sites during 1956 - 1960.
 - Airborne particulate activity averaged 1 particle/1000 ft³ above the 1 particle/1000 ft³ average found at the remote sites during 1956 - 1960.





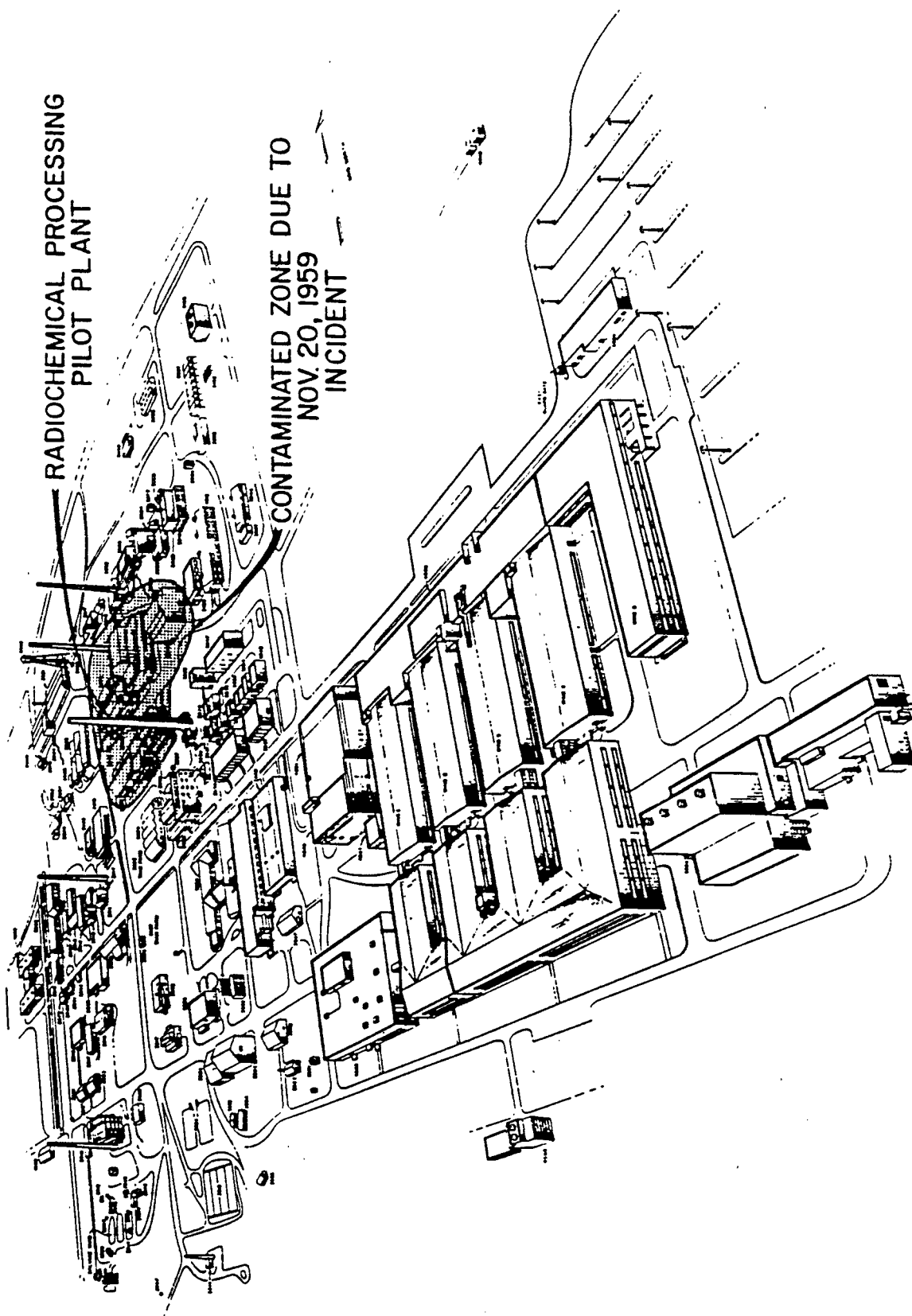


Fig. 4 Oak Ridge National Laboratory, X-10 Site.

Ref. 21

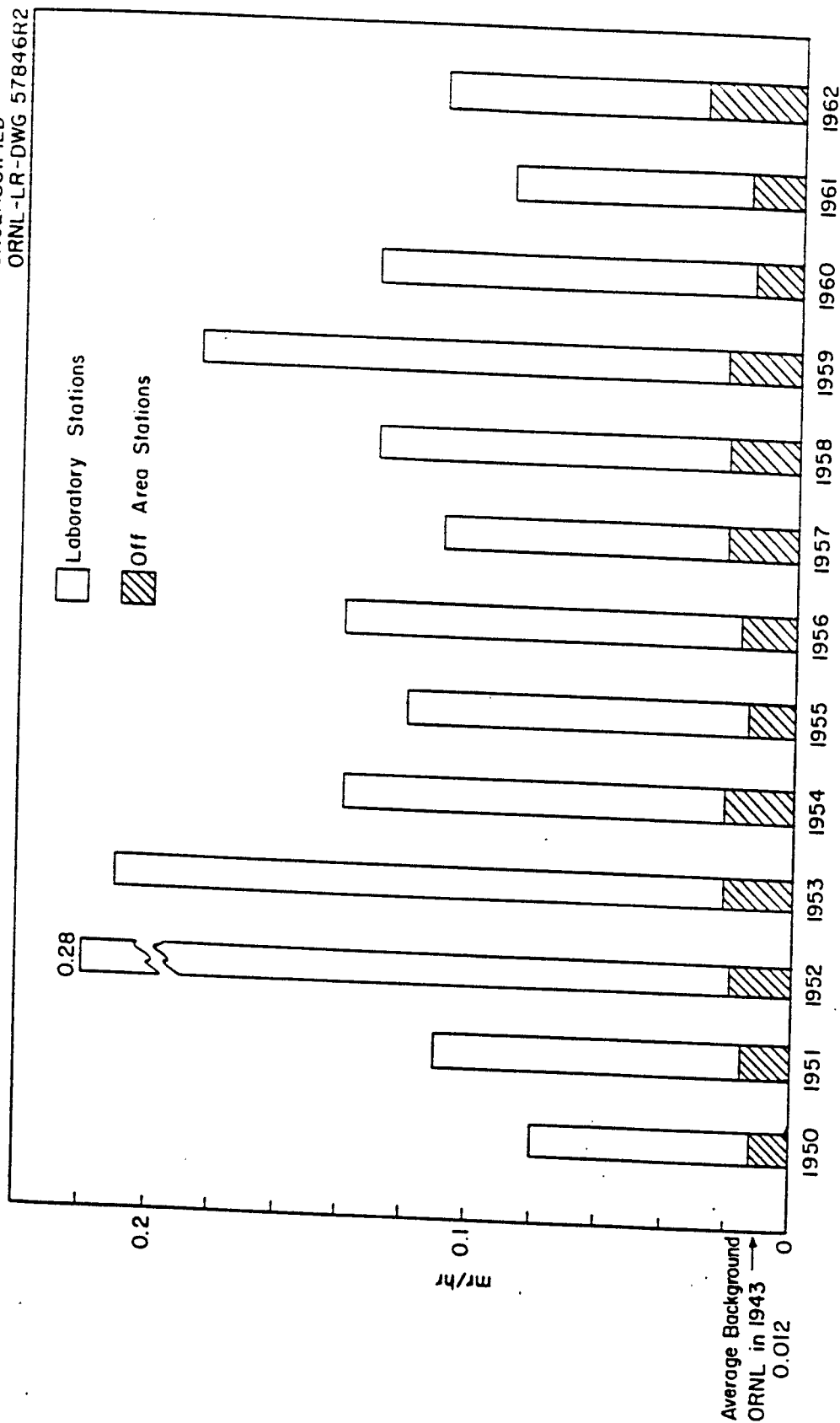


Fig. 5 Radiation Measurements Taken 3 ft Above the Ground Surface at ORNL Compared with Like Measurements Taken Elsewhere within the AEC Controlled Area for the Years 1950-1962.
Ref. 19

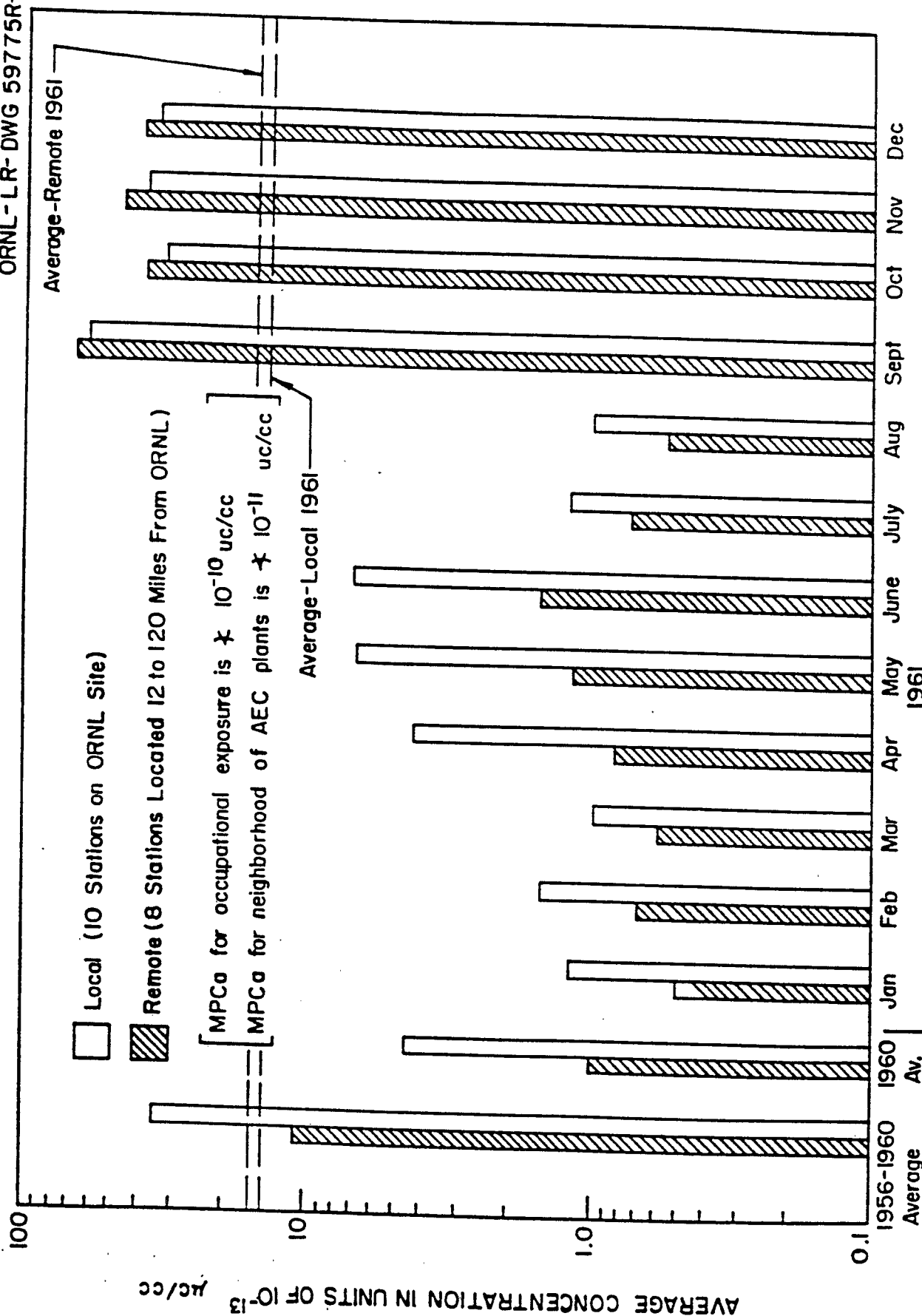


Fig. 6 Airbone Radioactivity Measurements (Filter Paper Data)
Ref. 20

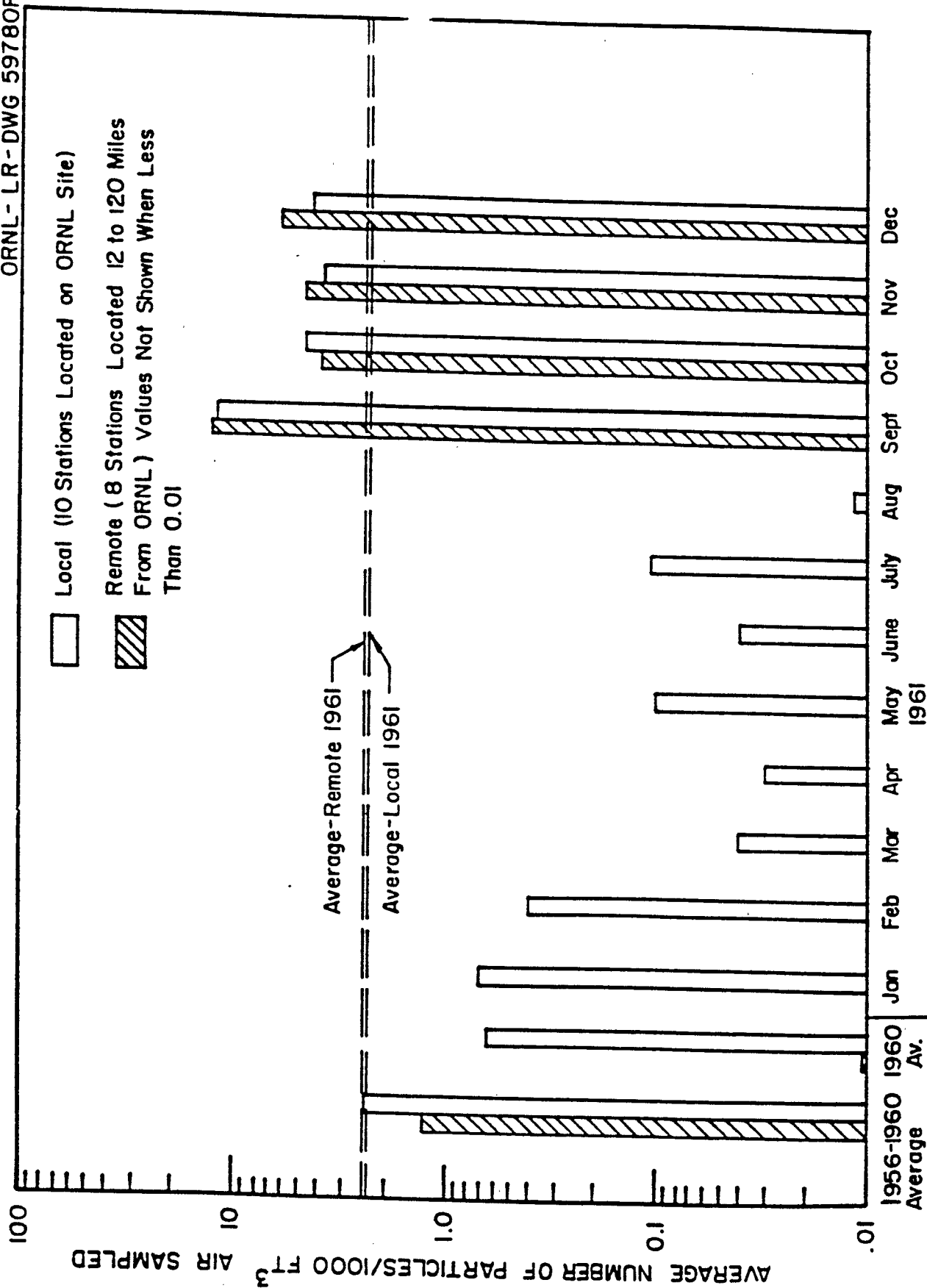


Fig. 7 Airborne Radioactive Particles Collected On Filters
(Measured by Autoradiographic Techniques)

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